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 FOREIGN DOCUMENTS OR RADIO BROADCASTS

REPORT

CD NO.

50X1-HUM

COUNTRY USSR

DATE OF
INFORMATION 1949

SUBJECT Scientific - Superconductivity

HOW
PUBLISHED Monthly periodical

DATE DIST. 27 Feb 1950

WHERE
PUBLISHED Moscow

NO. OF PAGES 3

DATE
PUBLISHED Mar 1949

LANGUAGE Russian

SUPPLEMENT TO
REPORT NO.

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SOURCE Zhurnal Eksperimental'noy i Teoreticheskoy Fiziki, Vol XIX, No 3, 1949,

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VARIATIONS IN THE SUPERCONDUCTIVE PROPERTIES
OF TANTALUM AS IT BECOMES SATURATED WITH HYDROGEN

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These experiments represent the first of a series in an attempt to determine, separately, the influence of absorbed gases and deformations upon the superconductive properties of tantalum. The first experiments, reported on in this article, demonstrated a very sharp change in the superconducting properties of tantalum specimens as they became saturated with hydrogen.

The tantalum specimens used for the study were wires 0.15 millimeter in diameter with a residual resistance $R_{4.2}/R_{290} = 0.2$. The wire served as a cathode in an electrolytic bath using distilled, slightly alkaline water. A cryostat was used to cool the specimen, after electrolysis, to the temperature of liquid helium in less than 2 minutes.

The temperature and magnetic curves of superconductivity transitions were recorded. The curves describing the dependence of the resistance of two tantalum specimens upon temperature for varying degrees of saturation with hydrogen show that the interval required for the superconductivity transition increased continuously toward lower temperatures as the specimen became saturated with hydrogen until finally the specimen lost its superconducting properties (at any rate, it was not superconducting down to 1.85 degrees K). For example, a specimen saturated with hydrogen until its resistance was approximately 1.3 times its normal (unsaturated) resistance at 4.2 degrees K became superconductive at 4.0 degrees K while the same specimen saturated until its resistance was 1.6 times the normal resistance did not become superconducting until approximately 3.05 degrees K. At the same time, however, the point marking the superconductivity transition still remained. A slight drop in resistance was observed at the transition temperature of the original unsaturated tantalum specimen, even for a specimen saturated with hydrogen until superconductivity disappeared. The residual resistance ($R_{4.2}/R_{290}$) of the most highly saturated

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specimen was three times as great as that of the unsaturated tantalum. A curve of the superconductivity transition for a tantalum specimen annealed in a poor vacuum (oxygen and nitrogen thus absorbed by the tantalum) shows that, although the transition temperature is lowered considerably, the transition remains sharp, in contrast to the elongated transition curves of the specimens saturated with hydrogen.

The transition curves for the breakdown of superconductivity by a longitudinal magnetic field again, as in the temperature transition curves, show unusual elongation of the transition. The transition interval required reaches hundreds of gauss and continues to expand as the metal becomes saturated with hydrogen.

Determination of the transition temperature and the critical field naturally becomes difficult as a result of the extreme elongation of both temperature and magnetic transitions. The generally accepted definition of these quantities as the temperature and field at which resistance becomes equal to half the residual resistance becomes quite unsuitable in this case.

The variations in superconductive properties of tantalum obtained when saturated with hydrogen can be linked with the amount of hydrogen absorbed by the metal. The determination of this amount is of course very difficult. However, an ideal example of the effect of this amount was given by the following experiment: a specimen, saturated with hydrogen until superconductivity disappeared and then annealed for 10 hours in a high vacuum at a temperature of 1,700 degrees, fully regained its superconductive properties; however, when this same specimen was again saturated with hydrogen, its superconductivity was lost when a much smaller amount of hydrogen was evolved on the specimen than was necessary before annealing. The amounts were approximately 340×10^{-3} milligram before annealing and only approximately 5×10^{-3} after annealing.

Next, an attempt was made to solve the problem of whether the hydrogen absorbed by tantalum forms compounds with the metal. Opposite views on this problem are held by Mary Andrews (Journ. Am. Chem. Soc., 54, 1845, 1932) and G. Hagg (ZS. phys. Chem., B, 11, 433, 1930). The results of these experiments were correlated with a previous work by K. Kan (Dissertation, Khar'kov, 1948) on the effect of all-sided compression upon the superconducting properties of metals to negate the possibility of hydrogen merely mechanically expanding the tantalum lattice. If, as Kan showed, a change in the parameter (coefficient of linear expansion) of the lattice by approximately 0.2 percent reduced the transition temperature by 0.006 degree under all-sided compression, then, if saturation with hydrogen only mechanically elongated the crystal lattice, an increase in transition temperature should be expected. The experiments, of course, showed the reverse effect; the transition temperature was lowered as the tantalum specimen became saturated with hydrogen. What actually takes place is the formation of a continuous series of alloys with hydrogen rather than the formation of definite compounds.

The fact that the critical field varies little as tantalum is saturated with hydrogen might indicate that small islands of tantalum unsaturated with hydrogen are the superconductors in this process. However, if this were true, high degrees of saturation should cause high values for the critical field by eliminating the islands, a condition which was not observed in the experiments. Saturation of tantalum with hydrogen does not cause marked inhomogeneities of the lattice; this would also cause a substantial increase in the critical field.

Thus, the experiments seem to prove that the superconductive properties of tantalum are extremely sensitive to absorbed gases. Webber's conclusion that the latter has little effect upon tantalum's superconductive properties (Phys. Rev., 72, 1241, 1947; Chem Abs, 42, 4, 1097, 1948) is therefore erroneous.

A secondary function of the experiments was the study of the influence of electrolytic saturation with hydrogen on niobium, which was used for superconducting electrodes in these experiments. It was shown that, under prolonged

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(many hours) saturation with hydrogen, niobium's superconductivity at 4.2 degrees K was retained. The opposite effect on the superconducting properties of such chemically closely related metals as niobium and tantalum must be studied in order to understand their general properties.

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